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(54) Abstract Title
Curable compositions

(57) A curable composition with an enhanced drying window for use in a primary or secondary imaging system is obtainable as the reaction product of an anhydride polymer and at least an ethylenically unsaturated material having one or more hydroxy groups. The anhydride functional polymer is obtainable as the reaction product of an ethylenically unsaturated monomer and an ethylenically unsaturated cyclic anhydride capable of copolymerizing with the unsaturated monomer. The latter reaction is effected in the presence of a free radical initiator with a low hydrogen abstracting capability. This is defined as an initiator having a hydrogen bond dissociation energy of no more than 430 KJ/mole. Suitable initiators are t-amyl peroxy compounds.

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CURABLE COMPOSITIONS

The present invention is directed to compositions which are thermally curable and/or curable by application of radiation, for example ultra violet (UV) radiation.

Such compositions are typically used in circuit board applications, either as etch resists, or when cured in combination with epoxy resins and the like, as solder resists. They also find application in deep chemical etching and graphic arts.

Typically, such materials are prepared by reaction of styrene with maleic anhydride in the presence of a free radical initiator. The resultant styrene/maleic anhydride resin is then reacted with hydroxy ethyl acrylate (HEA) or the like in the presence of a catalyst to produce the curable composition.

One problem with such curable systems is the drying window. The temperature at which the composition needs to be dried to enable clean development with aqueous alkali is lower than is optimally desirable. This problem has now been overcome by the present invention which allows the anhydride functional polymer to be prepared with increased linearity and a narrower molecular weight distribution. This manifests itself in a reduction in the polydispersibility and viscosity of the polymer. This is achieved by preparing the anhydride functional polymer using a free radical initiator which is a poor hydrogen abstractor.

Thus, the present invention now provides a curable composition obtainable as the reaction product of an anhydride functional polymer and at least one ethylenically unsaturated material having one or more hydroxy groups, the anhydride functional polymer being obtainable as the reaction product of at least one ethylenically

unsaturated monomer material with a cyclic anhydride material capable of reaction with the ethylenically unsaturated monomer material, in the presence of a free radical initiator which produces initiating radicals having a hydrogen bond dissociation energy of no more than 430 KJ/mole.

Curable compositions according to the present invention have greatly improved drying windows, relative to conventional systems, when used in a primary or secondary imaging system.

The compositions according to the present invention can be used on their own as etch resists. When used as solder resists, they may be presented in single pack form in which the composition is in admixture with an epoxy resin or the like, or they can be presented in a two-pack form in which the epoxy resin or equivalent is sold separately to be mixed with a composition of the present invention by the customer before use.

Initiators that produce radicals with a hydrogen bond dissociation energy of more than 430 KJ/Mole, for example methyl, t-butoxy, alkoxy and phenyl radicals are highly reactive and good hydrogen abstractors. These initiators produce anhydride functional polymers with a high polydispersivity.

Conversely, initiators that produce radicals with a hydrogen bond dissociation of 430KJ/Mole or less, such as ethyl, t-butyl and isopropyl radicals have now been found produce anhydride functional polymers with a much lower polydispersivity.

The hydrogen abstraction ability of the free radical initiator used in the preparation of the anhydride functional polymer can be expressed numerically in terms of its hydrogen bond dissociation energy which must be no greater than 430 KJ/mole. However, preferably, this bond dissociation energy is 420 KJ/mole or less, more preferably 410 KJ/mole or less.

For this use, the tertiary amyl peroxides are particularly preferred as free radical initiators. Mixtures of any of these alone or with one or more initiators of another type, whether or not also producing radicals with a hydrogen bond dissociation energy of 430KJ/mole may also be employed.

Other free radical initiators which have low hydrogen abstraction capability as indicated by a bond disassociation energy of no more than 430 KJ/mole are the azonitrile initiators, although sometimes these tend to produce products with higher levels of residual monomers and darker colours when compared with tertiary amyl peroxides. Mixtures of any of these alone or with one or more initiators of another type, whether or not also producing radicals with a hydrogen bond dissociation energy of 430KJ/mole may also be employed.

The ethylenically unsaturated monomer material which is reacted within the cyclic anhydride material is preferably selected from one or more (meth) acrylic and/or one or more vinyl monomers. Typically, suitable acrylate monomers be chosen from alkyl acrylates, typically having from 1 to 12 carbon atoms in acrylates, cycloalkyl acrylates typically having from 1 to 12 carbon atoms in the cycloalkyl group and other miscellaneous ether and other functional groups. In any acrylate of the foregoing and in any of the following specific examples of such materials, any acrylate group is optionally replaceable by methacrylate.

Suitable alkyl acrylates include acrylic acid, methyl acrylate, ethyl acrylate, n-propyl acrylate, iso-propyl acrylate, n-butyl acrylate, sec-butyl acrylate, tert-butyl acrylate, 2-ethylhexyl acrylate, hexyl acrylate, octyl acrylate, lauryl acrylate, isobornyl acrylate and dodecyl acrylate.

A typical cycloalkyl acrylate is cyclohexyl acrylate.

Suitable miscellaneous ether and other functional acrylates include di(ethyleneglycol) ethyl ether acrylate, di(ethylene glycol) 2-ethylhexyl ether acrylate, 2-methoxy ethylacrylate, 3-methoxy propyl acrylate and phenyl acrylate.

Mixtures of any of the foregoing may be employed, whether of the same class or different classes.

Suitable vinyl monomers may be aliphatic or aromatic. In the case of the aliphatic types, typical examples are vinyl acetate, vinyl chloride, vinyl propionate, vinyl pyrrolidone, vinyl versatate, vinylidene di chloride, vinyl fluoride, vinylidene di fluoride, 1-butene and 1-pentene etc.

Suitable aromatic vinyl monomers include styrene, vinyl toluene, α -methyl styrene, β -methyl styrene, 3-methyl styrene, 4-methylstyrene and vinyl naphthalene.

On the other hand, the cyclic anhydride which is reacted with the ethylenically unsaturated monomer material to produce the anhydride functional polymer may for example be selected from maleic anhydride, citraconic anhydride and itaconic anhydride and mixtures thereof.

Preferably, the relative amounts of the total ethylenically unsaturated monomer material and total cyclic anhydride material are in a weight ratio of the total ethylenically unsaturated monomer materials to the total cyclic anhydride of from 3:1 to 1:1.5, more preferably from 2:1 to 1:1.25, especially from 1.5:1 to 1.1:1, for example about 1:1.

The anhydride functional polymer preferably has an average molecular weight of from 700 to 100,000 when determined by the method of GPC analysis using a Perkin Elmer LC-30 RI Detector with a Perkin Elmer Liquid Chromatograph Series 10 Pump attached to a HPLC 360 Autosampler from Kontron, in conjunction with P.E. Nelson Systems Software Version 5.1. The GPC results are obtained by injecting 2% NVC solutions of the samples on to 1x30cm PLGel 10 μ mixed β linear column at 35°C with

a flow rate of 1ml/min analar tetrahydrofuran (THF). The calibration mix consists of polystyrene standards of molecular weights: 2750000, 220500, 11600 and 970. The molecular weight range of the column is 10000000 to 1000 and the GPC results are processed between these limits.

The anhydride functional polymer is reacted with at least one ethylenically unsaturated material having one or more hydroxy groups and this may be selected from a wide range of aliphatic or aromatic hydroxy functional(meth) acrylates. Suitable hydroxy functional acrylates (wherein the equivalent methacrylate is to be understood also to be recited) which may be employed, include 2-hydroxy ethyl acrylate, 3-chloro-2-hydroxypropyl acrylate, 2-hydroxy-1-methylethyl acrylate, 2-hydroxypropyl acrylate, 3-hydroxypropyl acrylate, 2,3-dihydroxypropyl acrylate, 2-hydroxybutyl acrylate and 4-hydroxybutyl acrylate.

To produce the final reaction product, as well as reacting the ethylenically unsaturated material having one or more hydroxy groups with the anhydride functional polymer, it is also possible to include in the reaction mixture, one or more amines and/or one or more saturated functional polyols and/or one or more imides with a polymerisable double bond. This offers the additional advantages of improvement in heat stability (particularly in the case of amines) and further widening of the pre-dry window (particularly in the case of polyols).

When one or more amines are included in the second stage reaction, these are preferably selected from one or more primary or secondary aliphatic or aromatic amines, for example mono-amines and diamines in which the or both amine groups (as appropriate) is attached to at least one substituent selected from C₁₋₆ (straight or branched) alkyl groups and phenyl groups, which alkyl or phenyl groups may themselves be optionally substituted by one or more non-reactive substituents. Typical amines for this purpose include ethylamine, diethylamine, propylamine, diaminopropane, ethylene diamine and butylamine.

The saturated functional polyol(s) can preferably be selected from one or more aliphatic or aromatic (including mixed aryl/alkyl type) polyalcohols, including straight or branched alkyl polyols having from 2 to 8 carbon atoms, and cyclic alkane polyols having from 2 to 8 carbon atoms. Typical examples are methanol, ethanol, propanol and cyclohexanol.

The imide(s) with a polymerisable double bond can be selected from maleimide, 3-maleimidopropionic acid, N-hydroxy succinimide ester, 4-(maleimidomethyl) -1-cyclohexane carboxylic acid, N-hydroxysuccinimide, N-methylmaleimide, N-ethylmaleimide and derivatives thereof, and mixtures of any of these.

In the second stage of reaction, the relative amounts of the anhydride functional polymer and the ethylenically unsaturated material having one or more hydroxy groups are preferably such that from 25 to 100 mole % of the anhydride groups, more preferably from 50 to 90 mole%, typically from 70 to 80 mole% are mono-esterified with the ethylenically unsaturated alcohol.

When saturated polyol(s) are included then their amounts relative to the anhydride functional polymer and the ethylenically unsaturated material are preferably from 5 to 70 mole%, more preferably from 10 to 50 mole%, especially from 15 to 35 mole%.

When amine(s) are also included, then their total amount relative to the anhydride functional polymer and the ethylenically unsaturated material having one or hydroxy groups are preferably such that from 2 to 50 mole%, more preferably from 5 to 30 mole% of the anhydride groups react with the amine(s).

A wide range of materials can be used for the ethylenically unsaturated material having one or more hydroxy groups which may be selected from one or more of hydroxy functional materials having a functional group capable of being cured on

exposure to radiation, e.g. UV light, for example hydroxy ethyl acrylate, hydroxyethyl methacrylate, hydroxypropyl acrylate, and mixtures thereof.

The second stage of reaction, i.e reaction of the anhydride functional polymer with at least the ethylenically unsaturated material having one or more hydroxy groups is preferably carried out in the presence of a catalyst, for example 1-methylimidazole, sodium acetate or lithium acetate, or mixture thereof.

In the two-pack form, the polymer can be cured by mixing with an epoxy resin containing at least two epoxy groups per molecule. Examples include novolac, bisphenol, triphenylmethane, as well as amino-copolymerised - and other epoxy resins.

However, in general, compositions according to the present invention may also comprise one or more optional ingredients such as photoinitiators, reactive diluents, inorganic fillers, colourants such as pigments or dyes and organic solvents.

Examples of suitable initiators include benzoin, benzoin methyl ether, 2,2-dimethoxy-2-phenyl acetophenone, 2-hydroxy-2-methyl-1-phenyl propan-1-one, acetophenone, 2,2-dimethoxy-2-phenylacetophenone, 1,1-dichloroacetophenone, benzophenone, methylbenzophenone, xanthane, 2-methylanthraquinone, 2-ethylanthraquinone, 1-chloroanthraquinone, 2-amylanthraquinone, 2,4-dimethylthioxanthone, 2-chlorothioxanthone, 2,4-diethylthioxanthone and 4-benzoyl-4'-methyldiphenyl sulphide, and mixtures thereof.

Accelerators such as tertiary amines or amine synergists, or thermal initiators such as azo or peroxide initiators, may be added to increase cross link density if required.

Examples of suitable colourants include phthalocyanine blue, phthalocyanine green, disazo yellow and crystal violet, and mixtures thereof.

Examples of suitable fillers include aluminium hydrate, china clay, calcium carbonate, micronised talc, barium sulphate, barium titanate, magnesium carbonate, calcium carbonate, aluminium carbonate and aluminium oxide, and mixtures thereof.

Examples of suitable reactive diluents include the acrylates or methacrylates of ethylene glycol, methoxytetraethylene glycol, polyethylene glycol, hexanediol, trimethylolpropane, pentaerythritol, ditrimethylolpropane, dipentaerythritol and trishydroxyethyl isocyanurate, and mixtures thereof.

Examples of suitable organic solvents include solvent naphtha, petroleum ether, petroleum naphtha, methyl ethyl ketone, cyclohexanone, toluene, xylene, ethyl acetate, butyl cellosolve acetate, carbitol acetate and propyleneglycol methylether acetate, and mixtures thereof.

The present invention will now be explained better by way of the following non-limiting examples.

Examples

Example 1

An anhydride functional polymer was first prepared by charging 206 grams of propyleneglycol methylether acetate to a reactor containing a mechanical stirrer, condenser, temperature probe and a nitrogen sparge, and heating to 143°C. A monomer mix was prepared consisting of 231 grams of propyleneglycol methylether acetate, 196.1 grams of styrene, 184.6 grams of maleic anhydride and 11.5 grams of di-tertiary amyl peroxide. The monomer mix was added to the solvent over a six hour period while maintaining the flask contents at 143°C. On completion of the addition the reaction mixture was then held for a further six hours at 138°C to ensure complete removal of the amyl peroxide. The anhydride functional polymer was found to have an

acid value of 275mgKOH/g and a viscosity at 40% N.V in propyleneglycol methylether acetate of 0.6 poise at 75°C. The mixture was then cooled to 85°C, an air sparge with a flow rate of 1/kg/hr applied and 0.6 grams 1-methylimidazole, 0.3 grams of hydroquinone and 163.9 grams of hydroxyethyl acrylate were charged. The mixture was held for 7 hours at 85°C then 6 grams of methanol was charged and the mixture held for a further 2 hours prior to discharge. This resin was called Resin 1.

Example 2

138 grams of aromatic hydrocarbon, Solvesso 150 ex Exxton, and 65 grams of carbitol acetate were charged to a reaction vessel and heated to 160°C. A monomer mix was then prepared with 228 grams of carbitol acetate, 193.4 grams styrene, 182.2 grams of maleic anhydride and 11.4 grams of tertiary amyl peroxy-2-ethylhexanoate. This was added to the reaction vessel maintaining an even rate over 5 hours while maintaining the temperature at 160°C. When the addition was completed, the contents of the reactor were cooled to 140°C and held for a further five hours. The anhydride functional polymer was then cooled to 90°C, an air sparge with a flow rate of 11/kg/hr applied and 3 grams of sodium acetate, 0.5 grams of hydroquinone and 161.7 grams of hydroxyethyl acrylate were added. The mixture was held for 5 hours at 90°C and 16.8 grams of propyleneglycol methylether added, after a further 3 hours at 90°C the product was discharged. The resin was called Resin 2.

Example 3

186 grams of propyleneglycol methylether acetate was charged to a reaction vessel and heated to 135°C. A monomer mix consisting of 223 grams of propyleneglycol methylether acetate, 188.7 grams of styrene, 177.7 grams of maleic anhydride and 11.0 grams of di-tertiary amyl peroxide was then added over 3 hours. The temperature being maintained at 135°C throughout the addition. When the addition was completed, the polymer was maintained at 135°C for a further 8 hours prior to cooling to 80°C. At 80°C an air sparge was applied at a flow rate of 11/kg/hr and 0.6 grams of

hydroquinone, 2.0 grams of 1-methyl imidazole and 211 grams of hydroxy ethyl acrylate added. The mixture was held for a further 12 hours at 80°C and then discharged. The resin was called Resin 3.

Example 4

203.2 grams of propyleneglycol methylether acetate was charged to a reactor and heated to 108°C. 231 grams of propyleneglycol methylether, 196.1 grams of styrene, 184.6 grams of styrene and 14.3 grams of tertiary-butyl peroxy-2-ethylhexanoate were then added at a uniform rate over six hours. The temperature was maintained at 108°C throughout the addition. On completion of the monomer addition the reaction mix was then held for a further six hours at 108°C. The anhydride functional polymer was found to have an acid value of 272mgKOH/g and a viscosity at 40% N.V. in propyleneglycol methylether acetate of 12.6 poise at 75°C. The mixture was then cooled to 85°C and an air sparge applied with a flow rate of 11/kg/hr, prior to charging 0.3 grams of hydroquinone, 163.9 grams of hydroxyethyl acrylate and 0.6 grams of 1-methylimidazole. The mixture was then held for 7 hours and 6 grams of methanol added, after a further 2 hours at 85°C the mixture was discharged. The resin was called Resin 4.

Four primary imaging resists were made to the formulation shown below, using a respective one of resins 1-4. Each ink was then coded 1-4 dependent on which resin it was made from.

		wt %
Resin	Resins 1-4	79.20
Monomer	TMPTA	4.00
Photoinitiator	Irgacure 907	4.00
Photoinitiator	Quantacure ITX	1.50
Filler	Talc IT Extra	4.00
Flow Aid	Byk 55	0.50

hydroquinone, 2.0 grams of 1-methyl imidazole and 211 grams of hydroxy ethyl acrylate added. The mixture was held for a further 12 hours at 80°C and then discharged. The resin was called Resin 3.

Example 4

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Four primary imaging resists were made to the formulation shown below, using a respective one of resins 1-4. Each ink was then coded 1-4 dependent on which resin it was made from.

		wt %
Resin	Resins 1-4	79.20
Monomer	TMPTA	4.00
Photoinitiator	Irgacure 907	4.00
Photoinitiator	Quantacure ITX	1.50
Filler	Talc IT Extra	4.00
Flow Aid	Byk 55	0.50

Dye	Crystal Violet	0.30
Solvent	Downanol PGDA	6.50

Inks 1-4 were then screen printed onto 0.5oz copper panels using a 77T/cm mesh and held for 5 minutes. Each board was then dried for 25 minutes at 115°C giving the dry film weights shown below:

Ink Number	Film Thickness, microns
1	10
2	10
3	11
4	9

A phototool and a 21 step stouffer wedge was then placed on each board. Each board was then exposed with 225mj of radiation through a Colight 5KW UV exposure unit, and developed with 0.6% sodium carbonate solution at 35°C. The results produced shown below:

Ink Number	Development	Solid Stouffer
1	Clean	9
2	Clean	9
3	Clean	11
4	Residue*	Residue*

*Ink residue remained on the board after development of the unexposed areas - processing was discontinued.

Boards were then alkali etched, rinsed and stripped as follows:

Etch	Chemistry	NH ₄ OH at 52°C, 8.5 pH
	Speed	230cm/min through 2 chambers
	Nozzle pressure	31 & 32 psi (U1 & U2), 26 & 25 psi (L1 & L2)
Rinse	4 Chamber cascade at 'high pH'	
Stripper	Chemistry	Atotech RR9F

Nozzle pressures 24 & 23 psi (U1 & U2), 25 & 25 psi (L1 & L2)

The final boards contained track widths of 50 and 75 microns, with only very minimal undercut.

In the light of this disclosure, modifications of the described Examples, as well as other Examples, all within the scope of the present invention as defined by the appended claims, will now be apparent to persons skilled in the art.

CLAIMS:

1. A curable composition obtainable as the reaction product of an anhydride functional polymer and at least an ethylenically unsaturated material having one or more hydroxy groups, the anhydride functional polymer being obtainable as the reaction product of an ethylenically unsaturated monomer material and a cyclic anhydride material capable of reacting with the ethylenically unsaturated monomer material, in the presence of a free radical initiator which produces initiating radicals having a hydrogen bond dissociation energy of no more than 430KJ/mole.
2. A composition according to claim 1, wherein the bond dissociation energy is 420KJ/mole or less, preferably 415KJ/mole or less.
3. A composition according to either preceding claim, wherein the free radical initiator comprises a tertiary amyl peroxide.
4. A composition according to any preceding claim, wherein the free radical initiator comprises an azo compound.
5. A composition according to any preceding claim, wherein the ethylenically unsaturated monomer material is selected from (meth) acrylate monomers, vinyl functional monomers and mixtures thereof.
6. A composition according to any preceding claim, wherein the ethylenically unsaturated monomer material is selected from alkyl acrylates, cycloalkyl acrylates, ether functional acrylates and other functional acrylates and methacrylic analogues of any of these and mixtures thereof.
7. A composition according to any preceding claim, wherein the ethylenically unsaturated monomer material is selected from aliphatic vinyl monomers, aromatic vinyl monomers and mixtures thereof.

8. A composition according to any preceding claim, wherein the cyclic anhydride material comprises a material selected from maleic anhydride, citraconic anhydride, itaconic anhydride and mixtures thereof.
9. A composition according to any preceding claim, wherein the anhydride functional polymer has an average molecular weight of from 700 to 100,000.
10. A composition according to any preceding claim, wherein the ethylenically unsaturated material having one or more hydroxy groups is selected from aliphatic and/or aromatic hydroxy functional (meth) acrylates and mixtures thereof.
11. A composition according to any preceding claim, wherein the final reaction product is obtainable as the reaction product of the anhydride functional polymer, the ethylenically unsaturated material having one or more hydroxy groups and one or more amines and/or one or more non-acrylated functional polyols and/or one or more imides with a polymerisable double bond.
12. A composition according to claim 11, wherein the amine(s) is/are selected from primary and/or secondary alkyl and/or aryl amines.
13. A composition according to claim 11 or claim 12, wherein the amine(s) is/are selected from ethylamine, diethylamine, propylamine, diaminopropane, ethylene diamine and butylamine and mixtures thereof.
14. A composition according to any of claims 11-12, wherein the non-acrylated functional polyol(s) is/are selected from alkyl, amyl, cycloalkyl, alkylanyl and arylalkyl poly-alcohols and mixtures thereof.

15. A composition according to any of claims 11-13, wherein the non-acrylated functional polyol(s) is/are selected from methanol, propanol, cyclohexanol and mixtures thereof.
16. A composition according to any of claims 11-14, wherein the imides is/are selected from maleimide, 3-maleimidopropionic acid, N-hydroxysuccinimide ester, 4-(maleimidomethyl)-1-cyclohexanecarboxylic acid, N-hydroxysuccinimide, N-methylmaleimide, N-ethylmaleimide and derivatives thereof, and mixtures thereof.
17. A composition according to any preceding claim, wherein the ethylenically unsaturated material having one or more hydroxy groups is selected from those which are capable upon exposure to radiation, especially U.V. light.



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Claims searched: 1-17

Examiner: Alan Kerry
Date of search: 21 May 1998

Patents Act 1977
Search Report under Section 17

Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:
UK Cl (Ed.P): C3J JCJ; C3P PHD
Int Cl (Ed.6): C08F 4/34; G03F 7038, 7/039
Other: Online databases: WPI, CLAIMS

Documents considered to be relevant:

Category	Identity of document and relevant passage	Relevant to claims
A	WO 91/03496 A1 (ISP) - see Claims 1-3, Examples 1 & 2 and pages 6 & 7	
A	US 4777230 (Kamath) - see Claim 3 and Example 1	

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.